Topological competition of superconductivity in Pb/Ru/Sr_2RuO_4 junctions

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We devise a new proximity junction configuration where an s-wave superconductivity and the superconductivity of Sr_2RuO_4 interfere with each other. We reproducibly observe in such a Pb/Ru/Sr_2RuO_4 junction with a single Pb electrode that the critical current I_c drops sharply just below the bulk T_c of Sr_2RuO_4 and furthermore increases again below a certain temperature below T_c. In order to explain this extraordinary temperature dependence of I_c, we propose a competition effect involving topologically distinct superconducting phases around Ru inclusions. Thus, such a device may be called a "topological superconducting junction".

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Despite extensive studies during the past several decades the realization of spin-triplet superconductivity has not been thoroughly established. Among a number of the candidates, the layered superconductor Sr_2RuO_4[1] is a most promising case for spin-triplet pairing.[2] The invariant spin susceptibility across the superconducting transition, observed by both NMR and polarized neutron scattering, provides indeed strong evidence for equal-spin pairing.[3,4] The odd parity nature of the orbital wave function, required for triplet states, is compatible with observation using the π-junction SQUID experiment.[5] There has been accumulated evidence to support the spin-triplet scenario.[6–8] For the complete confirmation of the superconducting parity of Sr_2RuO_4, however, it is highly desirable to conduct alternative direct experiments for the parity determination.

An important experiment aiming to detect the parity of Sr_2RuO_4 was introduced in 1999.[9] It was revealed that the critical current I_c of Pb/Sr_2RuO_4/Pb junctions with an s-wave superconductor Pb is sharply suppressed just below the superconducting transition temperature of Sr_2RuO_4, $T_{c,SRO} = 1.5$ K, and increases again at lower temperatures. The anomalous temperature dependence of I_c was interpreted as due to the change of *the phase difference between the TWO Pb electrodes* from 0 to π, driven by the odd parity superconductivity of Sr_2RuO_4.[10,11] In this study, we report similar behavior but using Pb/Ru/Sr_2RuO_4 junctions with essentially different configuration containing only a single Pb electrode. Thus we newly interpret the anomalous I_c as due to the change of *the winding number between Pb and Sr_2RuO_4*. Such behavior has never been reported in other superconductor/normal-metal/superconductor (SNS) junctions with an even-parity spin-singlet superconductor or those with odd-parity candidates UPt_3 or UBe_{13}.[12,13] The present finding provides evidence for a most intriguing interplay between the superconductors Pb and Sr_2RuO_4 connected via Ru metal inclusions, reflecting the distinct pairing symmetry of the two systems.

An SNS junction provides an opportunity to examine the quantum interference involving Sr_2RuO_4, since

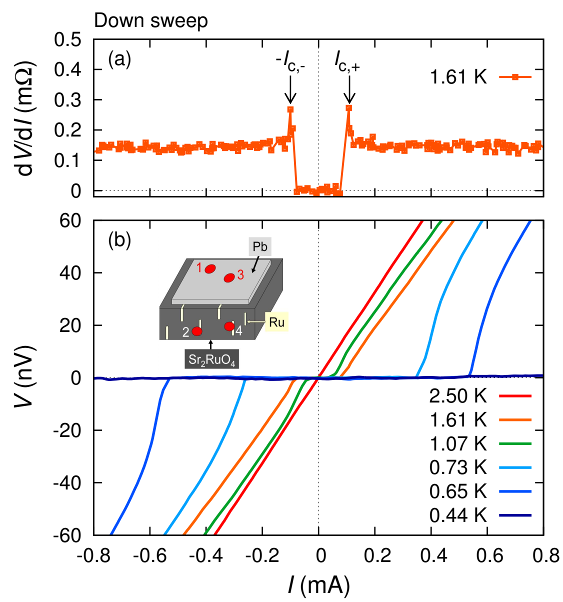

FIG. 1. (color online) (a) Differential resistance vs. current, and (b) voltage-current characteristics of a Pb/Ru/Sr_2RuO_4 junction. The inset to (b) is a schematic of a junction. The critical current $I_{c,+}$ is defined at the peak with positive bias current; $-I_{c,-}$ with negative bias current.

such interference sensitively affects its voltage-current (V-I) characteristics. Technically, poor electrical contact to the surface parallel to the basal ab-plane of a Sr_2RuO_4 crystal hampers the fabrication of a high-quality normal-metal/Sr_2RuO_4 junction. A Ru metal inclusion in a eutectic Sr_2RuO_4-Ru crystal[14] serves as an ideal normal-metal electrode, because of its atomically regular interface.[15] This eutectic system exhibits higher onset T_c of up to 3.5 K than $T_{c,SRO}$ ($= 1.5$ K) and T_c of Ru (~ 0.5 K), thus called the 3-K phase.[16] A number of experiments have revealed that the enhanced superconductivity occurs in the Sr_2RuO_4 region around Ru lamellae and possesses the same parity as that of Sr_2RuO_4.[17,18]

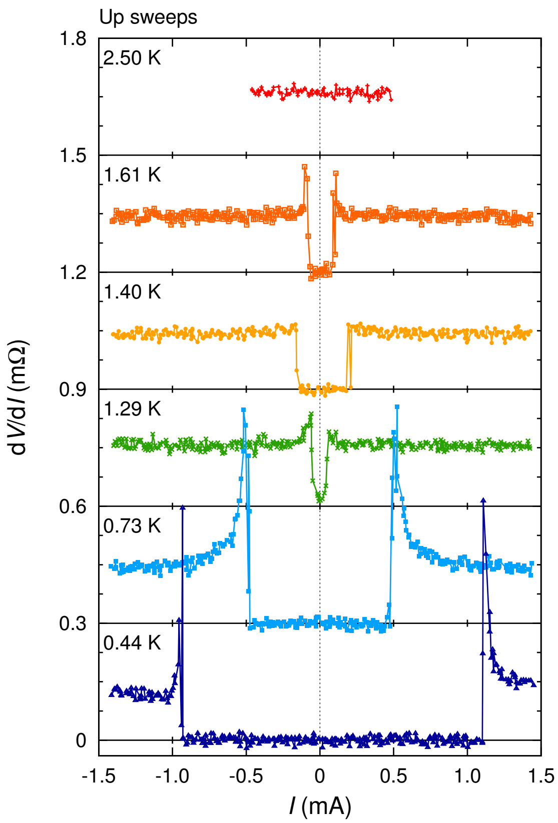

FIG. 2. (color online) Differential resistance $\mathrm{d}V/\mathrm{d}I$ of a Pb/Ru/Sr_2RuO_4 junction at various temperatures. Each curve is shifted by 0.3 mΩ for clarity. The critical current drops sharply below $T_{c,\mathrm{SRO}}$.

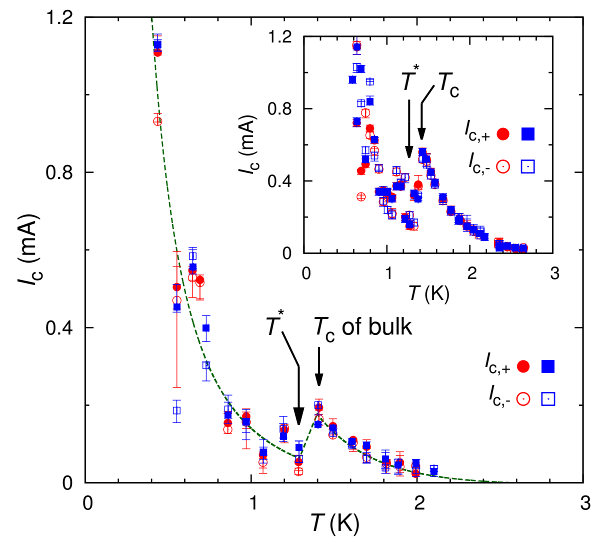

FIG. 3. (color online) Variation of the critical current I_c with temperature of a Pb/Ru/Sr_2RuO_4 junction. The inset displays I_c of another Pb/Ru/Sr_2RuO_4 junction. The circles and the squares indicate critical currents of up sweeps and down sweeps, respectively. I_c sharply drops just below $T_{c,\mathrm{SRO}}$ but increases again below a certain temperature designated as T^*. The broken curve is a guide to the eyes.

The eutectic crystals were grown by a floating-zone method.[19] The *ab*-surface of the crystal cut into the dimension of $\sim 1.9 \times 2.5 \times 0.2$ mm^3 was polished with diamond slurry. The area of Ru inclusions at the surface is typically less than 1% of the total sample area. A 1-μm thick film was deposited onto the polished surface using 6N-pure lead. In order to obtain the V-I characteristics between Pb and Sr_2RuO_4 using a four-probe method, two terminals each were put on Pb and Sr_2RuO_4 as illustrated in the inset to Fig. 1(b).

The differential resistance $\mathrm{d}V/\mathrm{d}I$ of Pb/Ru/Sr_2RuO_4 junctions was measured by applying an AC current at a frequency of 887 Hz using a lock-in amplifier. Fig. 1(a) is a representative curve of its dependence on the DC bias current at 1.61 K. Fig. 1(b) represents V-I characteristics at various temperatures obtained by integrating $\mathrm{d}V/\mathrm{d}I$, indicating the behavior of a typical superconducting junction. At each temperature, the data were taken after the junction was heated to 1.7 K and cooled to the measurement temperature with a negative DC current exceeding $-I_c$. At the target temperatures, the bias current was swept to a positive value (an up sweep) followed by a down sweep. All the curves in Fig. 1 represent the data taken on down sweeps. Fig. 2 represents the behavior in the up sweeps. As in Fig. 1(a), finite junction voltage emerges with a sharp peak in the differential resistance. Considering the amplitude of the AC current of 20 μA-rms, the critical current I_c is defined more precisely and accurately at the peak of $\mathrm{d}V/\mathrm{d}I$, rather than at the onset of non-zero $\mathrm{d}V/\mathrm{d}I$.

As shown in the top of Fig. 2, $\mathrm{d}V/\mathrm{d}I$-I curves are almost flat above 2.1 K. Below 2.1 K, the curves exhibit a dip-and-peak structure centered at the zero-bias current and the dip reaches zero at about 1.9 K, which is clearly higher than $T_{c,\mathrm{SRO}}$. I_c increases upon cooling below 2.1 K and the I_c-T curve in Fig. 3 exhibits a super-linear temperature dependence down to $T_{c,\mathrm{SRO}}$: $I_c \propto (T_c - T)^n$ with $n = 2.6$ for $T_c = 2.5$ K, or $n = 3.7$ for $T_c = 3.0$ K. For a tunneling junction with an insulator between the two superconductors, the Ambegaokar-Baratoff (A-B) theory gives $I_c = (eR)^{-1}\Delta_1 K([1-{\Delta_1}^2/{\Delta_2}^2]^{1/2})$ where R is the junction resistance in the normal state, $\Delta_{1,2}$ ($\Delta_1 < \Delta_2$) is the gap function of each superconductor, and $K(x)$ is a complete elliptic integral of the first kind.[20] For Δ_1 substantially smaller than Δ_2, as in the junctions studied here, the temperature dependence of I_c should actually be sub-linear, contrary to the behavior shown in Fig. 3. Furthermore, the I_cR values of our junctions, for example 0.01 μV at 1.4 K, are much smaller compared to an estimate of 400 μV using the A-B theory with $T_{c1} = 3$ K and $T_{c2} = 7$ K. These facts suggest that the Pb/Ru/3-K-phase configurations should be interpreted in terms of an SNS device with clean SN interfaces where the proximity effect induced coupling plays a crucial role.[21]

Just below $T_{c,\mathrm{SRO}} = 1.4$ K, used in the device represented here, the I_c suddenly drops to nearly zero. This

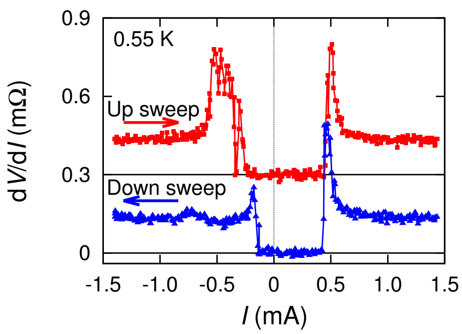

FIG. 4. (color online). Asymmetric and hysteretic bias current dependence of differential resistance $\mathrm{d}V/\mathrm{d}I$ of a $Pb/Ru/Sr_2RuO_4$ junction at 0.55 K. The up sweep curve is shifted by 0.3 mΩ. The down sweep curve was obtained immediately following the up sweep to 1.45 mA.

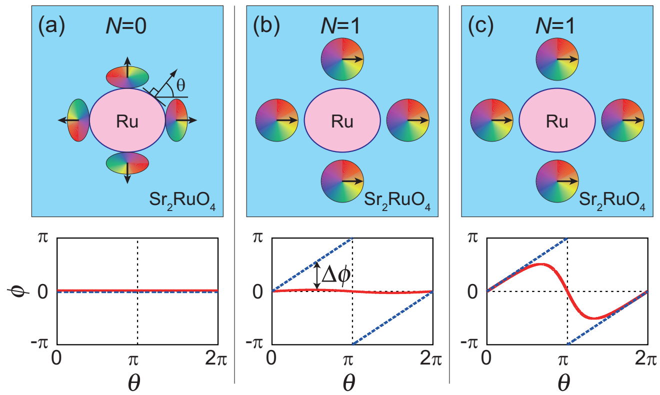

FIG. 5. (color online). Schematic images of the evolution of the order parameter at the Sr_2RuO_4/Ru interface. In the upper panels, the colors depict the momentum-directional dependence of the superconducting phase ϕ at each spatial position; the arrows denote the momentum direction for which $\phi = 0$. The angle θ is defined as normal to the interface (see (a)). The lower panels represent the superconducting phases $\phi(\theta)$ at the Sr_2RuO_4/Ru interface under no external current. The solid lines represent the phase of s-wave superconductivity in Ru and the broken lines that of p-wave superconductivity in Sr_2RuO_4. (a) $T_{\mathrm{c,SRO}} < T < T_\epsilon$: the $p_\parallel \pm i\epsilon p_\perp$ state with the winding number $N = 0$ is realized (A'-phase), matching with the s-wave order parameter induced in Ru. (b) $T \lesssim T_{\mathrm{c,SRO}}$: replacement by $p_x \pm ip_y$, the bulk state of Sr_2RuO_4, with $N = \pm 1$ (B-phase). (c) $T \ll T_{\mathrm{c,SRO}}$: increasing interfacial energy enlarges the phase deformation in the s-wave,[28] strengthening the Josephson coupling.

is a surprising result because below this temperature the interfacial superconductivity of the 3-K phase develops into the bulk superconductivity of Sr_2RuO_4 and under usual circumstances I_c is expected to increase rapidly. This distinct behavior marks clear evidence for unconventional interference effects. In addition, it indicates a qualitative change of the interfacial state of Sr_2RuO_4 at $T_{\mathrm{c,SRO}}$. Below $T_{\mathrm{c,SRO}}$, I_c remains suppressed in a narrow temperature range and exhibits complicated temperature dependence. As additional peculiar behavior, I_c starts to increase sharply below a certain temperature at about 1 K, which we designate as T^*. This anomalous overall temperature dependence is well reproducible among several samples. Another unusual feature of this junction is that the differential resistance as well as the associated I_c exhibit an unusual random variations below $T_{\mathrm{c,SRO}}$, as represented by the data points in Fig. 3.

In addition to the large variations in I_c, $\mathrm{d}V/\mathrm{d}I$ often becomes asymmetric with respect to the sign of the bias current below T^* as in Fig. 4. This asymmetric behavior implies the availability of many metastable order parameter configurations, similar to variable arrangements of the chiral p-wave domains in Sr_2RuO_4.[22–24]

Previous experiments and theories suggest that the 3-K phase originates most likely from the nucleation of superconductivity at the interface of Sr_2RuO_4 and Ru.[6] It corresponds to a single p-wave component existing in a narrow spatial range on the Sr_2RuO_4 side with its momentum parallel to the interface, denoted as $p_\parallel$-wave.[18] This time-reversal symmetry conserving state, called "A-phase", corresponds topologically to a superconducting state without phase winding ($N = 0$) from the viewpoint of the Ru-inclusion.[25] Below $T_\varepsilon = 2.4 - 2.6$ K the time-reversal symmetry breaking appears by adding an additional order parameter component, $p_\parallel + i\varepsilon p_\perp$,[18] which may correspond to the "A'-phase" with $N = 0$ or the "B-phase" with $N = 1$ within the theory introduced by Kaneyasu *et al.*[25] The latter is topologically compatible with the chiral p-wave bulk phase.

For the present junction geometry we assume that Pb induces superconductivity of s-wave symmetry in the Ru inclusions by proximity effect, which through spin-orbit interaction yields a direct coupling to the $p_\parallel$-wave order parameter in Sr_2RuO_4.[26,27] The topological matching with $N = 0$ of Ru naturally favors the A'-phase over the B-phase as depicted in Fig. 5(a), because the phase difference $\Delta\phi(\theta)$ at the interface can be set to zero for all angles θ around the circumference (junction ground state). While the A-phase consisting of only the $p_\parallel$-component is strongly localized at the interface, the A'-phase with the additional $p_\perp$-component is more extended (large normal-metal coherence length ξ_n perpendicular to the interface). This extension can explain why $I_\mathrm{c}(T)$ starts to increase slowly with lowering T below T_ε: the gradually growing order parameter $p_\perp$ strengthens the superconducting connections between different Ru-inclusions and to the external contacts. Thus, I_c is expected to grow approximately as $\exp[-d/\xi_\mathrm{n}(T)]$ with d the average connecting distance among Ru-inclusions and contacts and $\xi_\mathrm{n}(T) \propto (T - T_{\mathrm{c,SRO}})^{-1/2}$.[16] This yields behavior qualitatively compatible with the experimental results.

With the onset of bulk superconductivity at $T_{\mathrm{c,SRO}}$ the order parameter at the interface changes its topology to that of the B-phase (Fig. 5(b)), which due to its winding number $N = \pm 1$ is frustrated with the phase of the s-wave order parameter in Ru. For the over-

all Josephson current $I = \int_0^{2\pi} d\theta J_c(\theta,T)\sin\Delta\phi(\theta) \approx \bar{J}_c(T)\int_0^{2\pi} d\theta \sin\Delta\phi(\theta)$, this topological mismatch leads to an almost complete cancellation. At the same time, this frustration induces mild phase deformation just below $T_{c,\mathrm{SRO}}$. With decreasing temperature, the growing interfacial superconductivity in Sr_2RuO_4 requires less phase mismatch to minimize the total junction energy. As a result the region of significant $\Delta\phi$ is confined into a shrinking range of θ (Fig. 5(c)).[28] With the application of external current, the resulting phase deformation is such that the accompanying Josephson current density $J_c(\theta,T)\sin\Delta\phi(\theta)$ is constructively added and grows at lower temperatures.[29]

The extraordinary temperature dependence of I_c is explained in terms of a junction consisting of a topological superconductor encapsuling a conventional superconductor. For this reason, such a device may be called a "topological superconducting junction", in which nontrivial character of a topological superconductor becomes observable by appropriate design of the geometry.

The low-temperature phase ($T < T_{c,\mathrm{SRO}}$) introduces new features. One is the variation of the $\mathrm{d}V/\mathrm{d}I$-vs-I curves on the sweep direction of the supercurrent (see Fig. 4). This may involve complex phase frustration effects introduced below $T_{c,\mathrm{SRO}}$ through the phase winding of the B-phase, which can lead to various metastable states. Similar variations have been reported in different junctions of Sr_2RuO_4.[22,24] Since our present junctions contain a number of Ru inclusions acting as parallel contacts, future experiments using junctions consisting of a single Ru inclusion may resolve this issue.

The coupling of a Pb to Sr_2RuO_4 via a Ru-inclusion provides a unique opportunity to investigate quantum interference effects between topologically incompatible superconducting phases. The unusual sharp drop of the critical current I_c below $T_{c,\mathrm{SRO}}$ and its curious recovery below T^* can be explained by the change of the topology of the superconducting order parameter in Sr_2RuO_4 surrounding the Ru-inclusion. Thus, the device investigated here can be classified as a new class of superconducting junctions: a "topological superconducting junction".

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